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Examination Of Perlite-Polymer Interface Interactions in Polypropylene-Based Composites via Several Compatibilizers

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ABSTRACT

The surface interaction between the polymer and the mineral additive is one of the most significant aspects influencing the efficiency of mineral-filled polymeric composites. In this work, three distinct compatibilizers were introduced to composites based on polypropylene (PP) and perlite to improve interactions between the constituents. On composites comprising 10% expanded perlite content, three different ratios of ethylene vinyl acetate copolymer (EVA), thermoplastic polyurethane elastomer (TPU), and maleic anhydride grafted polypropylene (MA-PP) compatibilizers were employed. Composites were produced using an approach designated melt blending followed by injection mold-ing. The composites containing MA-PP compatibilizer possessed the most outstanding performance, according to the results of mechanical, physical, and dynamic mechanical evaluations and morphological characterizations. The investigated aspects indicated a rise in the composites containing 10 percent compatibilizer with the lowest adding amount, whereas performances declined at high compatibilizer contents. Along with these results, it was determined that the compatibilizers included in the PP/perlite composite system assisted in the fabrication of the composites by promoting the force values and melt flow rates identified during melt mixing. Following the test outcomes, MA-PP performed better than TPU and EVA in terms of compatibilizer efficiency. In general, it has been revealed that the selection of MA-PP compatibilizer in the manufacturing stages would offer benefits in terms of both simplicity of processing and mechanical strength where expanded perlite will be adopted as a natural filler for PP-based composites.

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Keywords:
Mineral additive; Polymer composites; Expanded perlite; Compatibilizer; Polypropylene; Polymer processing

INTRODUCTION

Because they are inexpensive and straightforward to handle, minerals are frequently used as reinforcement for polymer-based substances. Due to the low degree of incompatibility between the natural mineral surface and the polymer phase, the incorporation of mineral additives in polymeric composites has several limitations despite benefits like low weight, low cost, and recyclability (1-5). As a practical option to address the compatibility difficulties, tuning the chemical nature of the matrix-filler interface via integration of compatibilizer leads to improved mechanical resistance of composite materials (6-10).

The amorphous volcanic silica glass known as perlite is a naturally formed mineral with a high level of water. This aluminosilicate may expand by thirty times its original volume after heating. The majority of the world's perlite reserves, or more than 50%, are in Türkiye. Similar to volcanic particulate minerals, expanded perlite powder was used as an additive for various polymeric matrices (6, 11, 12). Polymers compounded with perlite mineral include polyolefins, polyesters, elastomers, and copolymers based on various forms such as films, foams, resins, fabrics, and 3D parts. Table 1 summarizes the polymers and investigated properties by citing these research studies (13-51).

As indicated in Table 1, research works dealing with PP/perlite composites were performed in four studies which reported that incompatibility between inor-

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Table 1. PER reinforced polymer composites in the literature.

Polymer matrix	Examined behavior	Reference number
Polyethylene (PE)	Thermal and mechanical	(13-19)
Polypropylene (PP)	Mechanical and processing	(20-23)
Polystyrene (PS)	Mechanical	(24, 25)
Polyvinylalcohol (PVA)	Thermal and mechanical	(26)
Acrylonitrile-butadiene-styrene (ABS)	Flame retardancy and mechanical	(27-29)
Polylacticacid (PLA)	Crystallinity and thermal stability	(30, 31)
Polyethyleneglycol (PEG)	Heat storage	(32)
Hydroxyapatite (HA)	Tissue engineering	(33, 34)
Polyaniline (PAn)	Electrical conductivity	(35)
Natural rubber	Odour-adsorbing	(36, 37)
Polyester fabrics	Acoustical	(38)
Polyphenylene sulphide (PPS)	Tribological	(39)
Polyurethane (PU)	Thermal insulation	(40-42)
Paraffin	Heat storage	(43, 44)
Epoxy resin	Ablative	(45-48)
Novalac resin	Tribological	(49)
Chitosan	Thermal stability	(50)
Polymethacrylicacid (PMAA)	Drug delivery	(51)
Butadiene rubber	Industrial goods	(52)

ganic perlite filler and organic PP phase influenced the mechanical behavior of composites negatively (20-23). Since the efficiency of maleic anhydride grafting is stated as the favored option for polymer composites (53-55), maleic anhydride grafted PP (PP-MA) was preferred as a compatibilizer. In addition to PP-MA, thermoplastic polyurethane (TPU) and ethyle-vinyl-acetate (EVA) were utilized to increase surface interactions in perlite-loaded PP composites attributed to their miscibility in the PP phase (56-59).

The interphase between additive and polymer phases plays a key role in the properties of composite materials attributed to physical and chemical aspects. The formation of interfacial interactions on the polymer-additive interface by integration of compatibilizers that can interact with both perlite and PP. For this reason, PP-MA, TPU, and EVA were compounded to enhance the interface adhesion of the inorganic perlite phase with organic PP matrix. Based on the findings of this study, performances of PP/perlite composites in terms of mechanical, morphological, processing, and melt-flow properties were compared to PP/EVA, PP/TPU, and PP/PP-MA composites involving perlite. Additionally, adding amount of perlite powder was investigated based on examined behaviors.

MATERIAL AND METHODS

Materials

PP with the trade name of Petoplen EH-251 was obtained from Petkim, İzmir, Türkiye. MA grafted PP was used as a compatibilizer with a commercial name Exxelor™ PO 1020 supplied by ExxonMobil, Texas, USA. The degree of grafting for PP-MA was indicated as a high graft level by the producer. Ester-based TPU (R130A85) was purchased from Ravago Petrochemicals, İzmir, Türkiye. The commercial name of EVA was Alcudia PA-461 supplied by Repsol S.A., Madrid, Spain. The vinyl acetate content of this EVA grade was 33%. According to their datasheets provided by producers, melting temperatures of EVA, TPU, and PP-MA polymers were 59 °C, 190 °C, and 162 °C, respectively.

Expanded perlite with a bulk density of 300–1000 g/cm^3 was supplied by Eti Maden, İzmir, Türkiye. The average particle size of PER was found to be 18 μm thanks to the particle size distribution curve shown in Fig. 1.

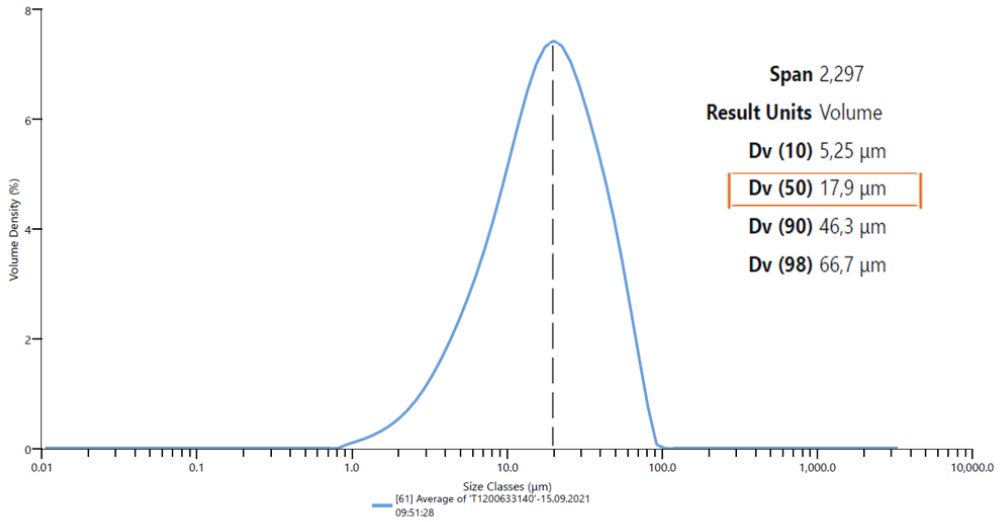

Figure 1. Particle size distribution curve of PER.

Composite Production

PP, TPU, PP-MA pellets, and PER powder were dried under vacuum at 80 °C for 2 h to remove moisture content before the compounding process. Composite samples were fabricated via Xplore MC15HT micro-extruder Adding amount of PER in composites was kept constant at 10% by weight. Three different concentrations for compatibilizers 10%, 20%, and 30% by weight were used. Processing temperature of 185°C, screew speed of 100 rpm, and mixing time of 4 min were applied during the melt-mixing process After the compounding step, dog-bone shaped test specimens with a dimension of 7.6 × 2.0 × 80 mm^3 for the tensile test and the dimension of 45 × 20 × 6.5 mm^3 for the D-type Shore hardness test was shaped using Daca injection molding instrument.

Characterization Techniques

Malvern Panalytical Mastersizer 3000 was used to evaluate the particle size of PER powder. Xplore Instruments program was employed to quantify force values throughout the extrusion process. The screw force values in the melt were determined using the micro-compounder's rheological software as a function of mixing time. Lloyd LR 30 K universal tensile testing machine was used for tensile properties of composites. Shore hardness values were determined using the Zwick R5LB041 digital hardness device. MFI measurements were performed via Coesfield Meltfixer LT using a 2.16 kg standard load at 185°C. The JSM-6400 Electron Microscope, a field emission scanning electron microscope, was used for observing the morphological characteristics of composite materials. A small coating of gold was applied to the surfaces of the cracked samples from the tensile test to establish conductive surfaces.

RESULTS AND DISCUSSION

Force Measurements

According to Fig. 2, force vs. time graphs demonstrate that PER additions elevated mixing force values due to powder incorporation boosted shear force as a result of increased melt viscosity throughout the extrusion operation. Compatibilizer inclusions with 10% content yielded lower force values compared to the PP/PER composite. The inclusion of EVA yielded a remarkable reduction in force values of PP/PER, whereas higher force values were obtained by TPU and PP-MA additions compared to the PP/PER EVA sample. Before high-scale production stages, this metric offers experimental data for planning manufacturing on the cost of fabricating the resultant composite materials. Since lower force data were recorded in compatibilizer-included composites relative to the force values of the PP/PER sample, the production of PER-filled PP composites can be carried out more economically in the presence of EVA, PP-MA, and TPU.

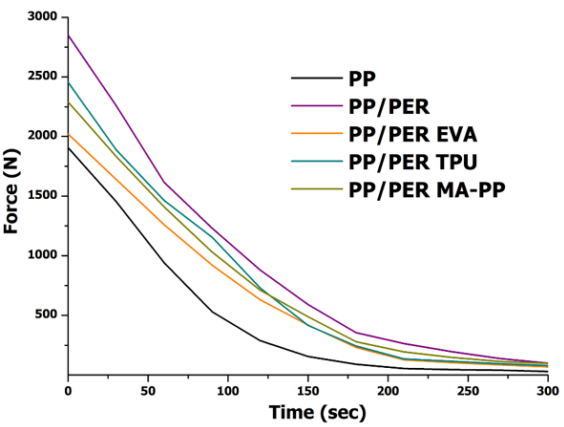

Figure 2. Force vs. time curves of PP and composites.

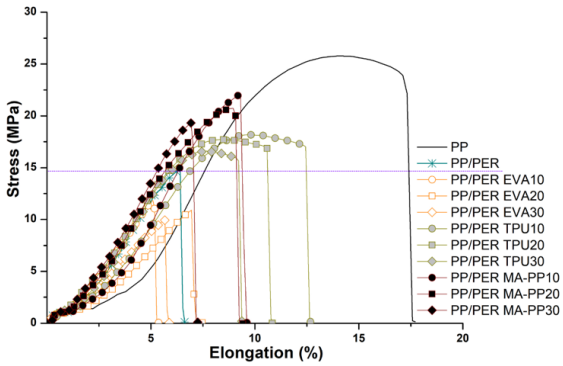

Figure 3. Stress vs. strain curves of composites.

Tensile Properties

Fig. 3 depicts tensile stress vs. percent strain curves. Tensile test data for PP and its composites, which include tensile strength, strain at break, and tensile modulus parameters are illustrated in Fig. 4.

The unfilled PP displayed ductile behavior according to its stress vs. strain curve in Fig. 3 in which necking behavior was observed. Since there was no necking property at the ultimate strength value. On the contrary, brittle characteristic was obtained after the incorporation of PER. Additionally, tensile stress and strain exhibited dramatic decline by PER addition. Using TPU as a compatibilizer caused to increase in the ductile behavior of composites stem from its high-strain elastomeric property whereas integration of PP-MA showed no effect on the brittle form of PP/PER.

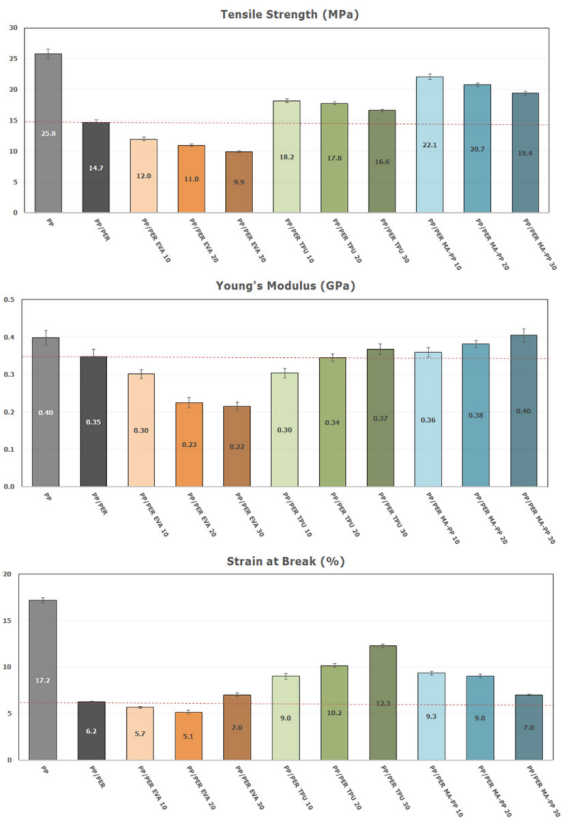

Figure 4. Tensile test data of samples.

Fig. 4 implied that EVA addition to the PP/PER system reduced tensile strength and Young's modulus parameters as well as strain values. As a common result, an increase in compatibilizer concentration resulted in a decline in the tensile strength of composites. TPU and MA-PP exhibited higher strength values compared to the PP/PER sample. The greatest tensile strength performance was achieved as PER-containing composites compounded with PP-MA compatibilizer since MA graft on PP structure enhanced compatibility between PER and polymer interface (55, 60).

Based on Young's modulus data, EVA displayed the worst performance among compatibilizers since remarkable reductions in modulus were observed. Conversely, TPU and PP-MA additions showed a positive effect on the modulus parameter of PP/PER. Young's modulus of composites enhanced as the added amount of compatibilizer increased

TPU-incorporated composites displayed a remarkable increase in percentage strain parameters attributed to the well-known high elongation behavior of TPU elastomer. PP/PER EVA composites gave the lowest results in terms of tensile strain.

Hardness Results

D-type Shore hardness data of unfilled PP and PER-filled PP composites are listed in Table 2. There were no significant differences in hardness measurement results compared to the reference material (PP). The shore hardness of PP displayed a slight increase with the addition of PER powder. Since EVA has an elastomeric nature, its

Table 2. Shore hardness results of composites.

Sample code	Hardness (Shore D)
PP	77.0 ± 0.1
PP/PER	77.5 ± 0.1
PP/PER EVA 10	76.5 ± 0.1
PP/PER EVA 20	76.0 ± 0.1
PP/PER EVA 30	75.0 ± 0.1
PP/PER TPU 10	78.0 ± 0.1
PP/PER TPU 20	78.5 ± 0.1
PP/PER TPU 30	79.5 ± 0.1
PP/PER MA-PP 10	77.5 ± 0.1
PP/PER MA-PP 20	77.5 ± 0.1
PP/PER MA-PP 30	78.0 ± 0.1

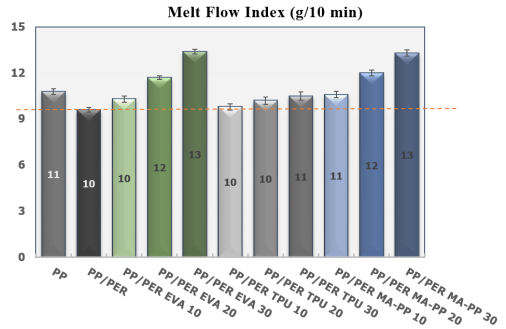

Figure 5. MFI data of samples.

integration into the PP/PER composite system resulted in reductions in the Shore D hardness of composites. On the contrary, TPU inclusions yielded improvement in hardness results despite it has elastomeric characteristics. The main reason for this observation might be the higher Shore D value of TPU with respect to PP. Similarly, PP-MA addition led to an increase in the Shore D parameter of the PP/PER composite.

Melt-flow Behaviors

Melt-flow index analysis is widely employed in thermoplastics for assessing the viscosity of molten polymers. Fig. 5 depicts the MFI characteristics of PP and related composites. When compared to unfilled PP, PER-incorporated composite possessed a lower MFI value. The rising quantity of MFI was found to be notable for composite samples containing a higher percentage of compatibilizers. TPU-loaded composites gave lower MFI values that were very similar to that of PP/PER. MFI specifications for composites were found to be in a limited spectrum while compared to unfilled PP as an overall finding regarding the melt-flow behavior of PER-loaded PP composites.

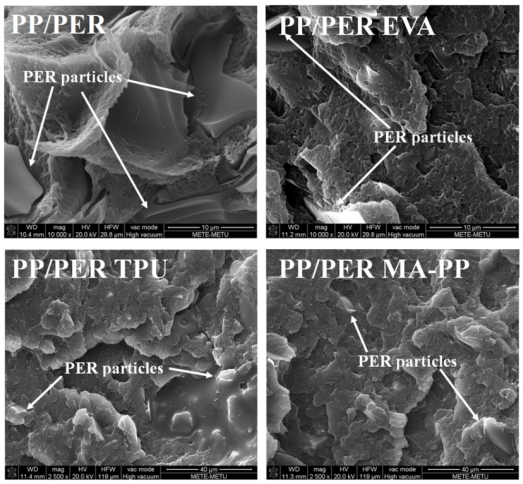

Figure 6. SEM micro-images of composites.

Morphological Analysis

The morphological study of composite samples was accomplished using SEM micro-images, which are illustrated in Fig. 6. Large gaps between PER particles and the PP matrix were detected in the SEM micrograph of PP/PER. The presence of compatibilizers donated strong adhesion and dispersion homogeneity in composite morphology. The formation of large gaps between phases was found to disappear after the inclusion of EVA as well as TPU. Similar to other compatibilizers, introducing PP-MA caused enhanced surface adhesion and distribution quality of PER according to the SEM image in Fig. 6. The result offered visible confirmation for improvements in associated composite performances pointed out in previous chapters.

CONCLUSION

One of the most crucial factors affecting the performance of mineral-filled polymeric composites is the surface interaction between the polymer and the mineral additive. In this research study, three different compatibilizers were used to enrich the interactions between phases in perlite-containing polypropylene (PP) composites. For this purpose, three different ratios of ethylene vinyl acetate copolymer (EVA), thermoplastic polyurethane elastomer (TPU), and maleic anhydride grafted polypropylene (MA-PP) compatibilizers were used on composites containing 10 percent expanded perlite mineral. Melt mixing followed by injection molding processes were used as the composite production process. In light of the findings obtained after mechanical, physical, and dynamic mechanical analysis and morphological characterizations, composites containing MA-PP compatibilizer showed the best performance. In the composites containing 10 percent compatibilizer with the lowest adding amount, an improvement was observed in the investigated properties, and a decrease in performances was detected at high compatibilizer concentrations. In addition to these findings, it was observed that the force values and melt flow rates measured during melt mixing, compatibilizers-introduced into the PP/perlite composite system facilitated the processing of the composites. According to the test results, the performance ranking among compatibilizers was found to be MA-PP > TPU > EVA. As a general conclusion, it has been evaluated that the preference of MA-PP compatibilizer in the production stages where expanded perlite will be used as a natural filler in PP-based composites will provide advantages in terms of both ease of processing and mechanical strength.

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CONFLICT OF INTEREST

Authors approve that to the best of their knowledge, there is not any conflict of interest or common interest with an institution/organization or a person that may affect the review process of the paper.

AUTHOR CONTRIBUTION

Çağrıalp Arslan: Methodology, Software, Validation, Writing- original draft. Ümit Tayfun: Data curation, Visualization, Investigation, Writing- review and editing. Mehmet Doğan: Supervision, Conceptualization.

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